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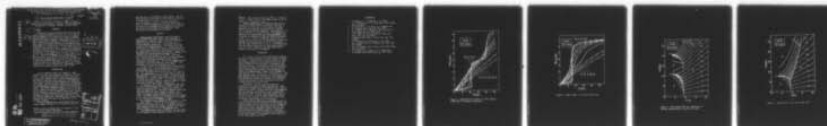
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6 FIELD-INDUCED TRANSITIONS IN DySb

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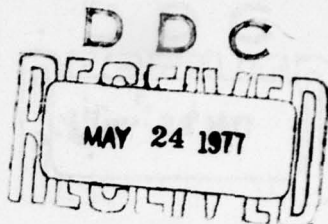
ABSTRACT

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The NaCl-structured compound DySb, which in zero field transforms abruptly at  $T_N \approx 9.5$  K to a Type-II antiferromagnetic (A) state with a nearly tetragonal lattice distortion, was previously found to exhibit rapid field-induced changes in magnetization at 1.5 K. The field-induced transitions in a DySb crystal have been studied by neutron diffraction and magnetization measurements in fields up to  $\sim 60$  kOe applied parallel to each of the principal axes. In the  $\langle 100 \rangle$  case, the transition from the A to an intermediate ferrimagnetic (Q) state is first-order at 4.2 K (critical field  $H_C \approx 21$  kOe) but is continuous from  $\sim 6$  K up to  $T_N$ , as  $H_C \rightarrow 0$ . The Q-to-paramagnetic (P) transition is rapid but continuous at 4.2 K ( $H_C \approx 40$  kOe) and becomes broad as  $T_N$  is approached. In the  $\langle 110 \rangle$  case the A-to-Q transition remains essentially first-order from 4.2 K ( $H_C \approx 15$  kOe) up to  $T_N$ ; above  $T_N$  rapid P-to-Q transitions occur at very high fields. The magnetic structure of the Q state is found to be that of HoP.

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INTRODUCTION

In zero magnetic field, the compound DySb exhibits a first order magnetic transition<sup>1,2</sup> at  $T_N \approx 9.5$  K. Above  $T_N$  DySb has the cubic NaCl-structure. Below  $T_N$  the compound orders antiferromagnetically in the type-II structure that consists of ferromagnetic (111) planes stacked antiferromagnetically along the [111] axis. The first order transition is accompanied by a predominantly tetragonal lattice distortion ( $c/a = .993$ ). The ordered magnetic moment at 6 K is  $9.5 \mu_B$  per Dy, almost the saturation moment of  $10 \mu_B$  for  $Dy^{3+}$ , and is parallel to the tetragonal [001] axis. Elastic as well as magneto-thermal data for DySb have been analyzed in a molecular field approximation,<sup>3,4</sup> and the analysis shows that biquadratic pair interactions between the  $Dy^{3+}$  ions may be as important as the bilinear exchange.

Previous magnetization measurements<sup>5</sup> made on a single crystal of DySb at 1.5 K in fields up to 60

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kOe show that a ferrimagnetic state exists. For all three principal directions of the field the values of the magnetization in this state are consistent with the magnetic structure of DySb being that of HoP,<sup>6</sup> in which the moments within each (111) plane of Dy atoms are ferromagnetically aligned with  $10 \mu_B$  per Dy and oscillate between two perpendicular  $\langle 100 \rangle$  directions in alternating (111) planes.

## RESULTS

Magnetization measurements were performed on single crystals of DySb using a vibrating-sample magnetometer. Magnetic fields of up to 56 kOe were applied parallel to the  $\langle 100 \rangle$ ,  $\langle 110 \rangle$  and  $\langle 111 \rangle$  crystallographic directions. The fields have been corrected for demagnetization. The results for the  $\langle 100 \rangle$  direction are shown in Fig. 1 as isotherms  $M$  vs  $H$ . Below  $T_N$  we observe two critical fields at which the magnetization increases very rapidly. At 4.2 K the magnetization changes discontinuously at  $H_{c1}^{\langle 100 \rangle} = 21.6$  kOe and very rapidly at  $H_{c2}^{\langle 100 \rangle} = 40.6$  kOe. Both critical fields decrease with increasing temperature, both transitions becoming broader as the temperature approaches  $T_N$ . At 4.2 K the magnetization is almost  $5 \mu_B$  per Dy atom just above  $H_{c1}^{\langle 100 \rangle}$  and approaches  $10 \mu_B$  per Dy atom above  $H_{c2}^{\langle 100 \rangle}$ .

In Fig. 2 the  $M$  vs  $H$  isotherms are shown for the field along  $\langle 110 \rangle$ . The isotherms show one transition which remains abrupt while its critical field,  $H_{c1}^{\langle 110 \rangle}$ , decreases from 15.6 kOe at 4.2 K to zero at  $T_N$ . The magnetization at 4.2 K approaches a value of approximately  $7.2 \mu_B$  at high fields. In addition, the isotherm for 9 K shows a very rapid increase in  $dM/dH$  at fields just above  $H_{c1}^{\langle 110 \rangle}$ . The 10 K and 12 K isotherms above  $T_N$  show a similar change in  $dM/dH$  at fields just above 15 kOe and 26 kOe, respectively.

To determine the ranges of temperature and field for the various magnetic states of DySb, we have converted our data to field vs temperature curves of constant magnetization. The phase boundaries have been identified from the rapid changes in  $dH/dT$ . Fig. 3 shows the magnetic phase diagram for  $H$  parallel to  $\langle 100 \rangle$ . The phase marked P is paramagnetic, the A phase is the Type-II antiferromagnetic state, and the Q-phase is the intermediate ferrimagnetic state.

The magnetic phase diagram for  $H$  parallel to  $\langle 110 \rangle$  is shown in Fig. 4. The P, A, and Q phases are the same as for  $H$  parallel to  $\langle 100 \rangle$ . An unusual feature of this diagram is that the boundary between the Q and P phases extends up to 15 K at the maximum field.

The magnetization data for  $H \parallel \langle 111 \rangle$  are not shown, but the magnetic phase diagram is similar to that for



$\vec{H} \parallel \langle 110 \rangle$ . The critical field,  $H_{cl} \langle 111 \rangle$ , is 19 kOe at 4.2 K, and the value of the magnetization in the Q phase is  $\sim 5.8 \mu_B$  per Dy atom.

Neutron diffraction measurements have been performed on a single crystal of DySb with  $\vec{H} \parallel [110]$  to determine the structure of the Q-phase. We have measured the integrated intensities of the  $(\frac{1}{2} \frac{1}{2} \frac{1}{2})$  and the  $(\frac{5}{2} \frac{3}{2} \frac{1}{2})$  antiferromagnetic reflections at 4.2 K and 11 K and for  $H = 0$  and 30 kOe. The observed intensities for the Q-phase are in good agreement with  $10 \mu_B$  per Dy parallel to  $[100]$  and  $[010]$  in alternate  $(111)$  planes. At 11 K no magnetic reflections are present in zero field, but by increasing the field the  $(\frac{1}{2} \frac{1}{2} \frac{1}{2})$ -reflection appears at the P to Q boundary, with the same intensity as at 4.2 K. Preliminary data for the lattice distortions in the Q phase indicate that the lattice expands in the  $[001]$  direction, normal to the plane of the moments.

#### DISCUSSION

The results of the present magnetization measurements are in good agreement with the results of Busch and Vogt.<sup>5</sup> As pointed out by these authors the value of the magnetization in the Q phase for each of the three principal field directions is consistent with the HoP structure. Furthermore if the three Q phases have identical magnetic structures with the same exchange and anisotropy energies, the differences between the Zeeman energies of the three Q-phases predict that  $H_{cl} \langle 100 \rangle = \sqrt{2} H_{cl} \langle 110 \rangle = 2/\sqrt{3} H_{cl} \langle 111 \rangle$ . The experimental values of the three critical fields are in good agreement with this prediction. Both magnetization and neutron diffraction results for the Q-phase of DySb show therefore that this phase has the HoP structure with  $10 \mu_B$  per Dy atom.

For  $\vec{H} \parallel \langle 100 \rangle$  the HoP structure cannot be stabilized by bilinear exchange interactions alone; additional couplings such as biquadratic pair interactions are needed. Stevens and Pytte<sup>7</sup> have suggested that the dominant contribution to this biquadratic interaction comes from the couplings between the ion and the lattice. In their model cooperative shifts of the Sb ions relative to the Dy ions create local distortions stabilizing the HoP structure.

Further neutron diffraction measurements are required to verify the HoP structure of the Q phases for other direction of the field and to test the model of Stevens and Pytte.<sup>7</sup> Our magnetization data for DySb above  $T_N$  are being compared in detail with crystal-field calculations of magnetization vs effective field in order to expose any evidence for biquadratic or any other higher-order coupling, as was done in a recent investigation of PrAg.<sup>8</sup>

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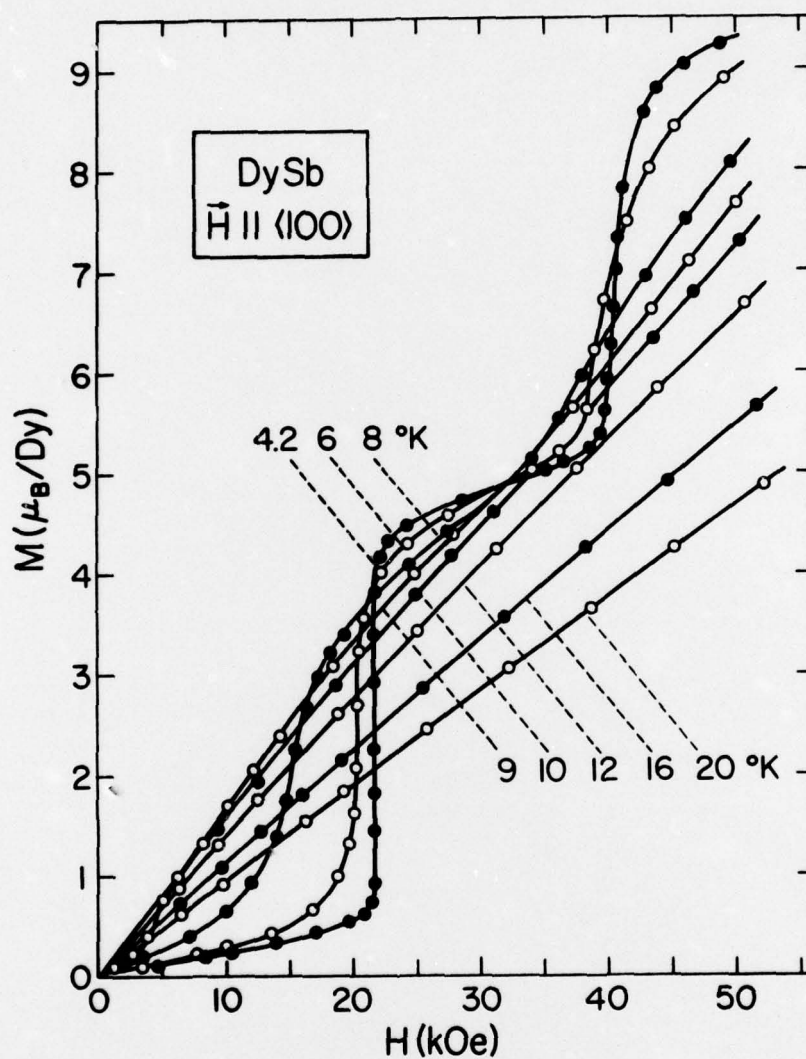


Figure 1. Magnetization of DySb vs. field applied along  $\langle 100 \rangle$  at constant temperature.



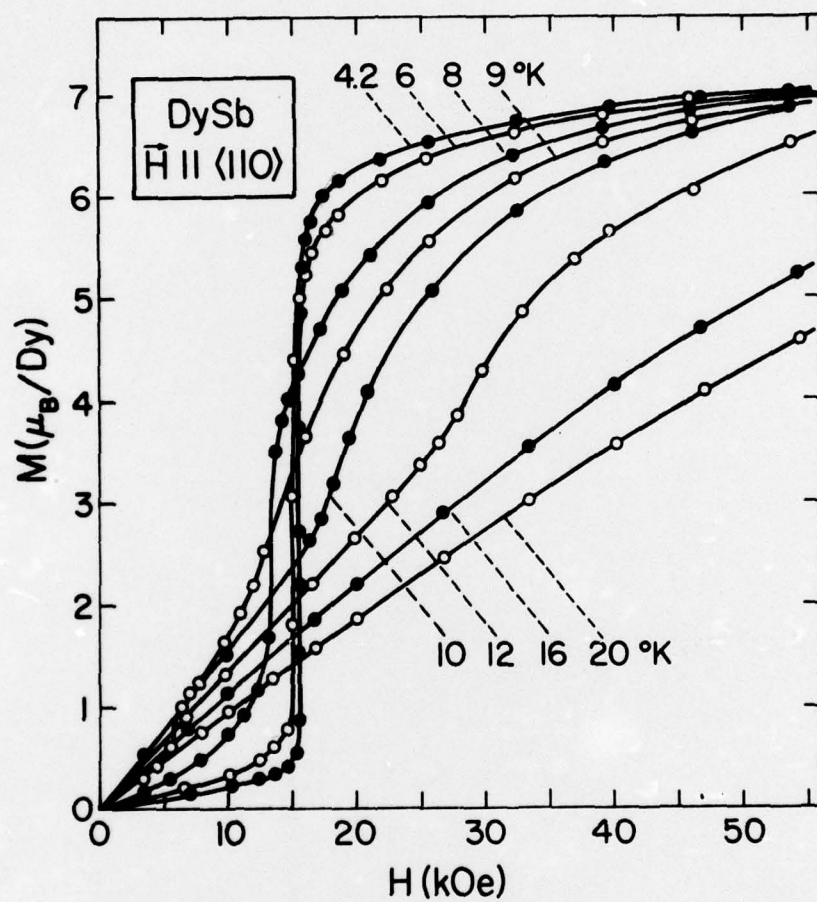


Figure 2. Same as Fig. 1 for field along  $\langle 110 \rangle$ .

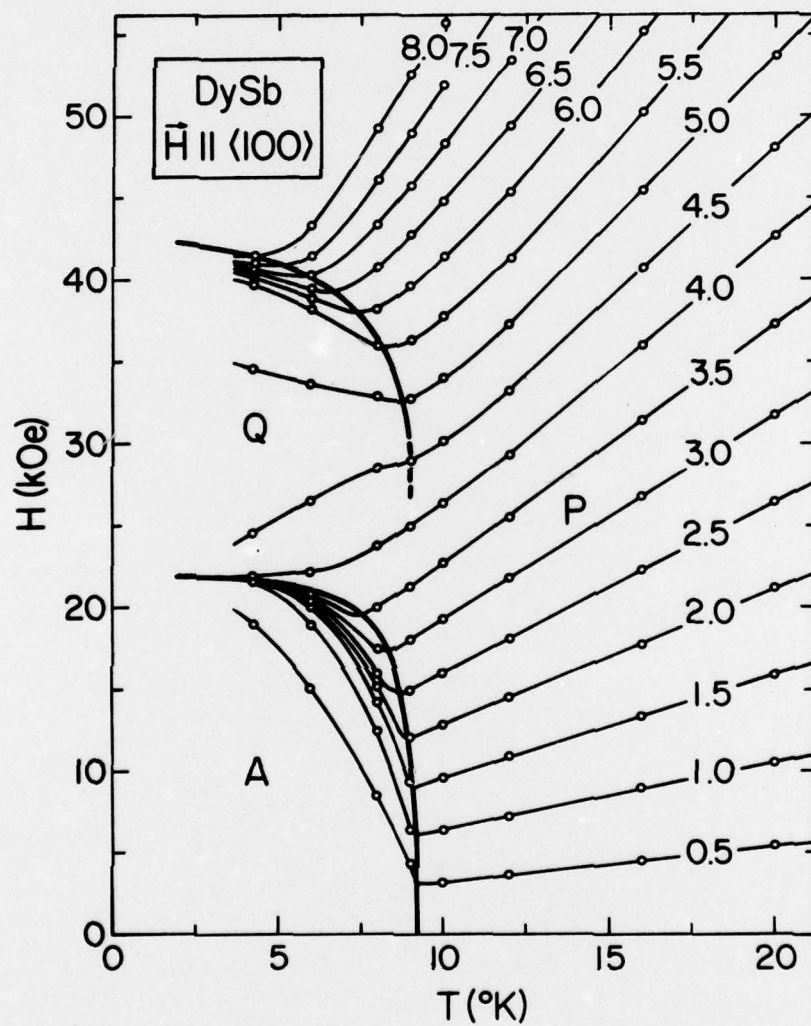


Figure 3. Field along  $\langle 100 \rangle$  vs. temperature at constant magnetization (in  $\mu_B/\text{Dy}$ ) for DySb.



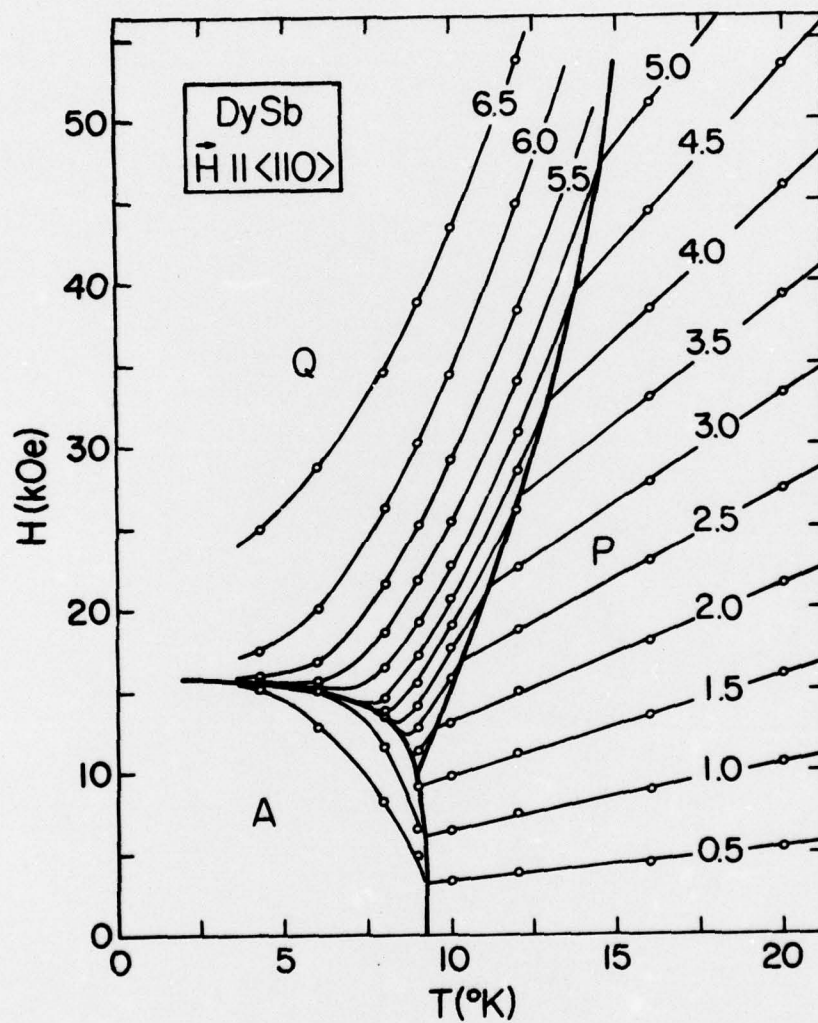


Figure 4. Same as Fig. 3 for field along  $\langle 110 \rangle$ .